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Synthesis of geopolymer from large amounts of treated palm oil fuel ash: Application of the Taguchi method in investigating the main parameters affecting compressive strength

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HIGHLIGHTS

- Geopolymer was synthesized from high content of treated palm oil fuel ash.
- TPOFA-based geopolymer mortar gained compressive strength of 47 MPa at 7 days.
- The optimized TPOFA-geopolymer mortar mixture was analysed using XRD and FTIR.
- The main binding phases consist of N–A–S–H and C–S–H gels.

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ABSTRACT

The aim of this study was to synthesize geopolymers using a large amount of treated palm oil fuel ash (TPOFA). The efficiency of the TPOFA (as the source material) in producing geopolymer products was enhanced via six factors which were optimized using the Taguchi method L25. The six factors were divided into two different components: (i) additive materials i.e. $Ca(OH)_2$, silica fume (SF), $Al_2(OH)_3$, and (ii) alkaline activators; i.e. NaOH concentration (moles), Na-silicate: NaOH ratio, and alkali-activator:solid-material ratio. Each of these factors was examined on five levels in order to obtain the optimum mixture. A total of 25 mixtures were prepared in accordance to the L25 array proposed by the method. The performance of the specimens was evaluated by compressive strength tests. The results show that the optimum mixture consisted of 65 wt.% TPOFA and 35 wt.% additive materials which achieved a compressive strength of 47.27 ± 5.0 MPa after 7 days of curing. The properties of the optimized mixture were further analyzed via X-ray diffractography (XRD) and Fourier transform infrared spectroscopy (FTIR) analyses. The results show that the main binding phases consist of aluminosilicate type gel "N–A–S–H" (Na_2O–Al_2O_3–SiO_2–H_2O) and calcium silicate hydrate (C–S–H) gels, formed simultaneously, within the TPOFA-based geopolymer mortar.

1. Introduction

A geopolymer is also known as an alkali-activated aluminosilicate material. A geopolymer binder has a three-dimensional amorphous structure of (N–A–S–H) type gel (Na_2O–Al_2O_3–SiO_2–H_2O). The gel is formed by either the polymerization of individual $[SiO_4]^{4-}$ and $[AlO_4]^{5-}$ species as building blocks in the system which are the main reaction product of alkali-activated aluminosilicate (geopolymer) materials derived from low-calcium content [1,2] or a calcium alumina silicate hydrate C–(A)–S–H type gel (CaO–Al_2O_3–SiO_2–H_2O) which is the main binding phase in the system of alkali-activated aluminosilicate calcium-rich source material [3,4]. Therefore, geopolymer synthesis is dependent on the use of material rich in aluminum–silicate glass activated with alkaline solutions [5]. The most commonly used material is calcined kaolin or metakaolin (MK) [2,6]. However, industrial wastes such as fly ash and ground granulated blast furnace slag (GGBFS) are also used due to their high silica and alumina content with little and high CaO content, respectively [1,3]. In the major palm oil producing countries, palm oil fuel ash (POFA) is another abundantly available waste material which can be used for geopolymer syntheses. POFA [7] has high SiO_2 (61.33) content, low Al_2O_3 (7.018) content, and high P_2O_5 (4.55) content compared to class C and class F fly ash, GGBFS, and MK or calcined kaolin [1–4,6]. It also has

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higher Fe_2O_3 (5.11) content compared to GGBFS, and MK or calcined kolin [2,3,6], and higher MgO (4.69) and CaO (8.20) content compared to class F fly ash and MK [1,2]. Theoretically, there should be a direct correlation between the geopolymerization results and the total silica content of the source materials. Increasing the amount of silica causes the number of Si–O–Si bonds which are stronger than Si–O–Al and Al–O–Al bonds [8], to increase; this implies that the strength of geopolymers would also increase with the Si/Al ratio because the density of the Si–O–Si bonds increases with the increase in Si/Al ratio [9,10]. Hence, the high silica content of the POFA reflects its feasibility as source material for geopolymer synthesis.

Untreated POFA has been used in combination with pulverized fuel ash (PFA) at a ratio of 30:70, and activated with an alkaline activator to produce geopolymer with a compressive strength of 30 MPa after 28 days of curing [11]. This rather limited strength potential of the resulting geopolymer could be attributed to the high content of unburned carbon in POFA, as unburned carbon of base material has been found to affect compressive strength of geopolymer [12]. Therefore, POFA has to be treated to reduce the carbon content in order to promote the geopolymerization process. The potential of POFA, especially treated POFA (TPOFA) [7], as a source material for geopolymer synthesis has not been adequately explored as in the case of class C and class F fly ashes, GGBFS, and MK or calcined kaolin [1–4,6]. Nonetheless, this is changing as great interest in POFA has sparked among researchers, especially those from palm oil producing countries.

To the authors' best knowledge, no geopolymerization of TPOFA has ever been reported, possibly due to its efficiency as it contains a high amount of silica, but a low amount of alumina, as well as calcium which need to be initially enhanced. Recent research works have shown that the inclusion of specific amounts of calcium hydroxide ($Ca(OH)_2$) [13], aluminum hydroxide ($Al(OH)_3$) [14], and silica fume (SF) [15] can enhance the geopolymerization processes. In all cases, they were done individually in different aluminosilicate source materials. A significant amount of these materials showed considerable impact on the geopolymerization processes. Therefore, it is important to introduce TPOFAs containing different amounts of these materials in order to investigate the enhancement of TPOFA for efficient geopolymer synthesis.

Another crucial parameter that affects the TPOFA geopolymer synthesis is alkali activator types (combination and concentration). Several works have been carried out on geopolymers, yet no clear explanation has been provided on the effects of various factors for upsetting source materials, such as how the strength increased with high NaOH concentrations [16,17]. However, some other researchers [18,19] found that high concentrations of NaOH have a negative effect on strength. Moreover, previous research has concluded that the use of Na_2SiO_3/NaOH in a weight ratio of 1.0 gives strength of up to 70 MPa [20]. The highest compressive strength value of 71 MPa was observed at 2.5 Na_2SiO_3/NaOH weight ratio [21].

Investigating all of these parameters ($Ca(OH)_2$ wt.%, SF wt.%, $Al(OH)_3$ wt.%, NaOH concentration (mole), Na_2SiO_3/NaOH (weight ratio), and alkali-activator/solid materials (weight ratio)) in a single work may not be possible. However, with a suitable design method, one may consider some of the factors affecting these properties. The Taguchi experimental design is one of the most famous methods used to design the parameters of specific problems. The application of the Taguchi method in geopolymers by past researchers for similar purposes [22–24], but with different source materials, has been successful.

This study is an investigation on the synthesis and characterization of geopolymers produced using TPOFA as the source material. Six design factors were examined at five levels by the Taguchi method to obtain the optimum mixture. The six factors are $Ca(OH)_2$ wt.%, SF wt.%, $Al(OH)_3$ wt.%, NaOH concentration (mole), Na_2SiO_3/NaOH (weight ratio), and alkali-activator/solid materials (weight ratio). A total of 25 experiments were conducted according to L25 array proposed by the Taguchi method. The efficiency of the TPOFA was enhanced by investigating the effects of the abovementioned parameters on the mechanical properties of the product in order to obtain the optimum mixture. This mixture was then examined using X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR).

2. Materials and methods

2.1. Materials

Four basic raw materials were used to produce the geopolymer mortar. Raw POFA was collected from a nearby palm oil mill in Nibong Tebal, Penang, Malaysia. The incompletely combusted fibers and kernel shells were separated using a 300 μm sieve. The POFA was then ground in a ball mill to obtain particle sizes of about 10 μm. To remove the unburned carbon, POFA was heated at 500 °C for 1 h in a gas furnace and then the POFA was subjected to second stage grinding to form the TPOFA. The chemical and physical properties of the TPOFA in this study are provided in Table 1. Based on ASTM C618 [25], the treated POFA could be classified as a class F mineral admixture. The same approach has recently been used and was reported to be effective in increasing the efficiency of TPOFA for use in high-strength green concrete [7] and engineered cementitious composites [26]. The TPOFA is the major source material in this research. The alkaline activators used were solutions combining NaOH and Na_2SiO_3. The analytical grade NaOH was in pellet form with 98% purity and the commercial Na_2SiO_3 was in liquid form with a specific gravity of 1.53 g/cm^3 at 20 °C and a silica modulus (Ms, where Ms = SiO_2/Na_2O) of 2; 14.7% Na_2O, 29.4% SiO_2, and 55.9% H_2O were combined. The additives used were $Ca(OH)_2$, $Al(OH)_3$, and SF. The surface area of $Ca(OH)_2$, $Al(OH)_3$ and SF were 0.6214 m^2/g, 0.3298 m^2/g, and 0.1364 m^2/g, respectively. Clean river sand passing a 1.18 mm sieve and retained on a 150 μm sieve, with a fineness modulus of 2.8 and specific gravity of 2.65, was used.

2.2. Design of the mixtures

In this study, the Taguchi method was used to design the mixtures and to obtain the optimum mixture design by considering the effects of the parameters on their mechanical properties. Six primary factors were examined followed by a statistical study of the $Ca(OH)_2$ wt.% (designated as A); the SF wt.% "B"; the $Al(OH)_3$ wt.% "C"; the NaOH concentration (moles) "D"; the Na_2SiO_3 to NaOH weight ratio "E"; and the liquid alkaline to solid material weight ratio "F". Each factor was examined at the five levels as described in Table 2. The level of each factor and the values of the tested factors were chosen based on previous researches [14,27–29]. The amount of water added to each mixture was calculated based on the percentage of geopolymer paste at 5% by weight ratio [30]. All of the geopolymer mortars were made with sand to solid material weight ratios of 1.5 which is similar to that used in previous researches [31,32]. This is the optimum ratio for the binder (solid material) and sand; anything beyond this ratio will cause the compressive strength to be dramatically reduced [33]. The design suggested by the Taguchi method for six factors at five levels is the L25 array, as shown in Table 3, while Table 4 shows the values and trial mixture proportions used in the series of 25 mixtures. The compressive

Table 1
Chemical compositions using XRF technique and physical properties of TPOFA.

Chemical	Component (%)
SiO_2	61.33
Al_2O_3	7.018
Fe_2O_3	5.11
CaO	8.20
MgO	4.69
P_2O_5	4.55
K_2O	6.50
SO_3	0.27
TiO_2	0.25
MnO	0.097
Na_2O	0.123
C	1
Physical properties	
Specific surface area (m^2/g)	1.775
Loss on ignition (%)	2.53
Median particle size d_{50} (μm)	2.06